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Polarizability of DNA Block Copolymer Nanoparticles Observed by Electrostatic Force Microscopy

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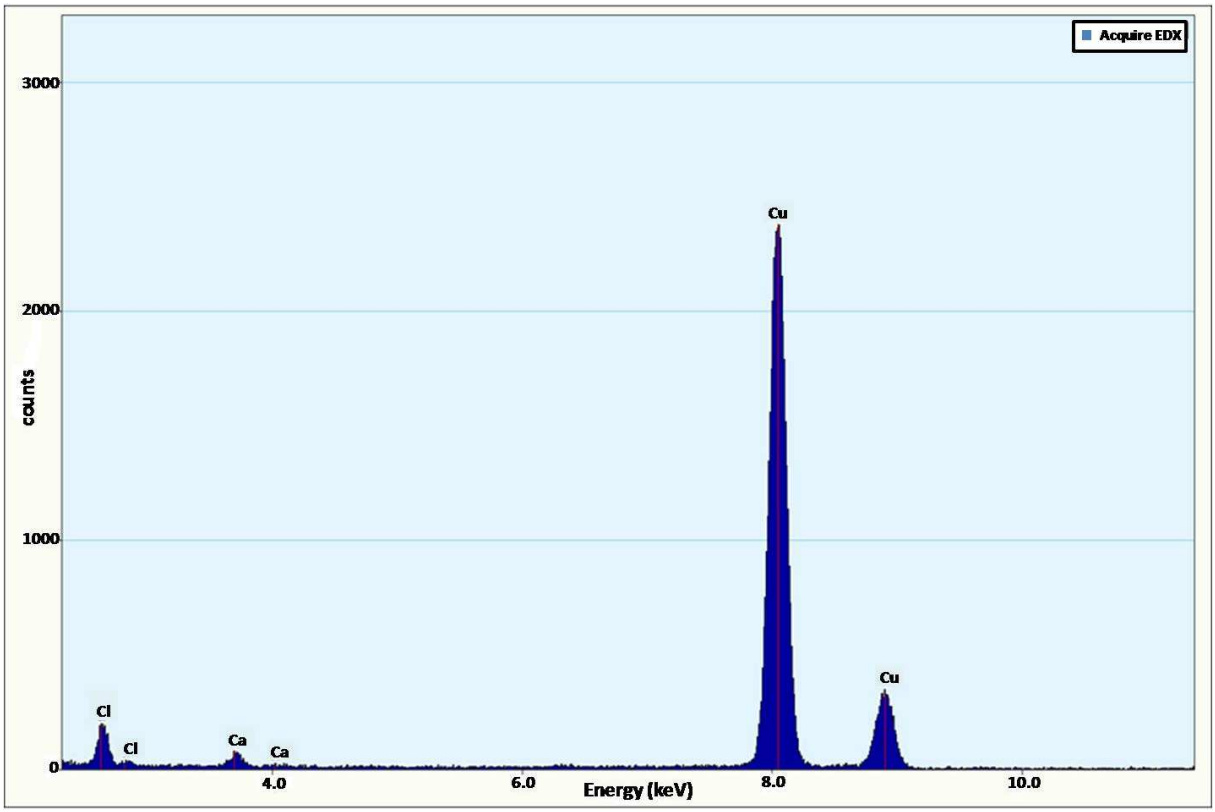

Figure S1: Energy Dispersive Microanalysis measurements (EDS) coupled to a High Resolution Transmission Electron Microscope (HRTEM) of non-doped DNA-*b*-PS micelles.

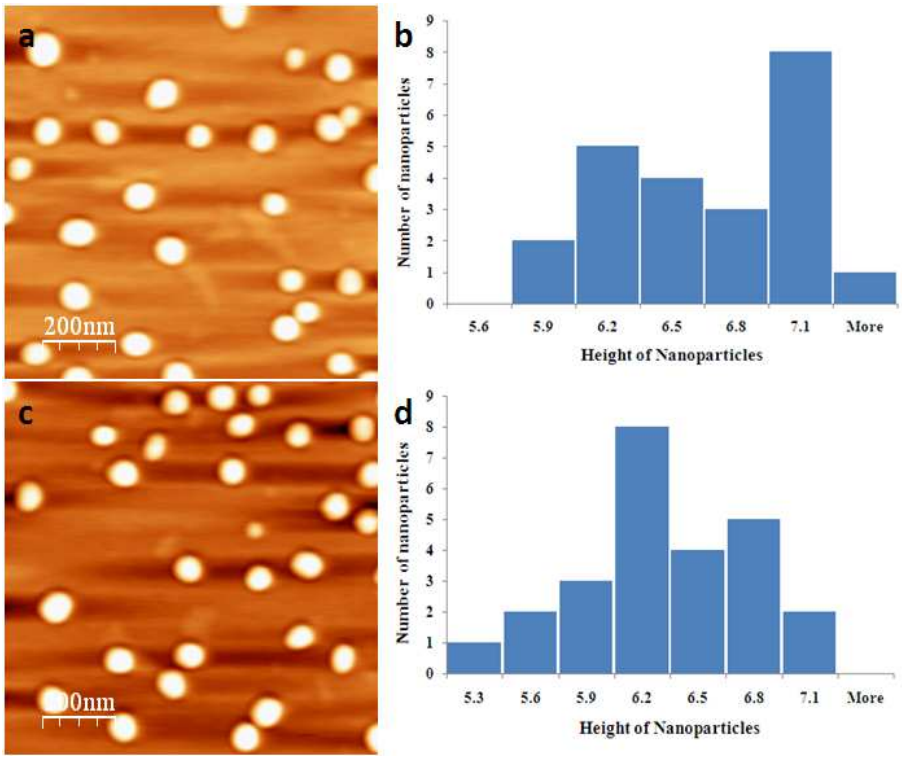

Figure S2: Atomic Force Microscopy (AFM) topography images of DNA-*b*-PS aggregates recorded in tapping mode. (a) Ferrocene-doped DNA-*b*-PS micelles deposited on a mica surface. (b) Height distribution of the doped micelles with an average height of 6.5 ± 0.4 nm. (c) Undoped micelles deposited on a mica surface. (d) Height distribution of the undoped micelles with an average height of 6.2 ± 0.5 nm.

AFM and EFM of ssDNA deposited on a mica surface. Sample preparation: A droplet of 10µL of ssDNA solution (conc. 20 nM) with the same sequence composition as that of the micellar corona was applied to a freshly cleaved mica surface for 5 minutes. The sample was rinsed gently with triple-distilled water and dried with nitrogen gas. The single-stranded 22mer ODN molecules appear in the AFM tapping mode images as small objects with a height of around 1 nm. The EFM measurements were acquired at 10 nm above the set point. Phase shift images (EFM) of the same area at 5 V, 0 V and -5 V show no EFM signal.

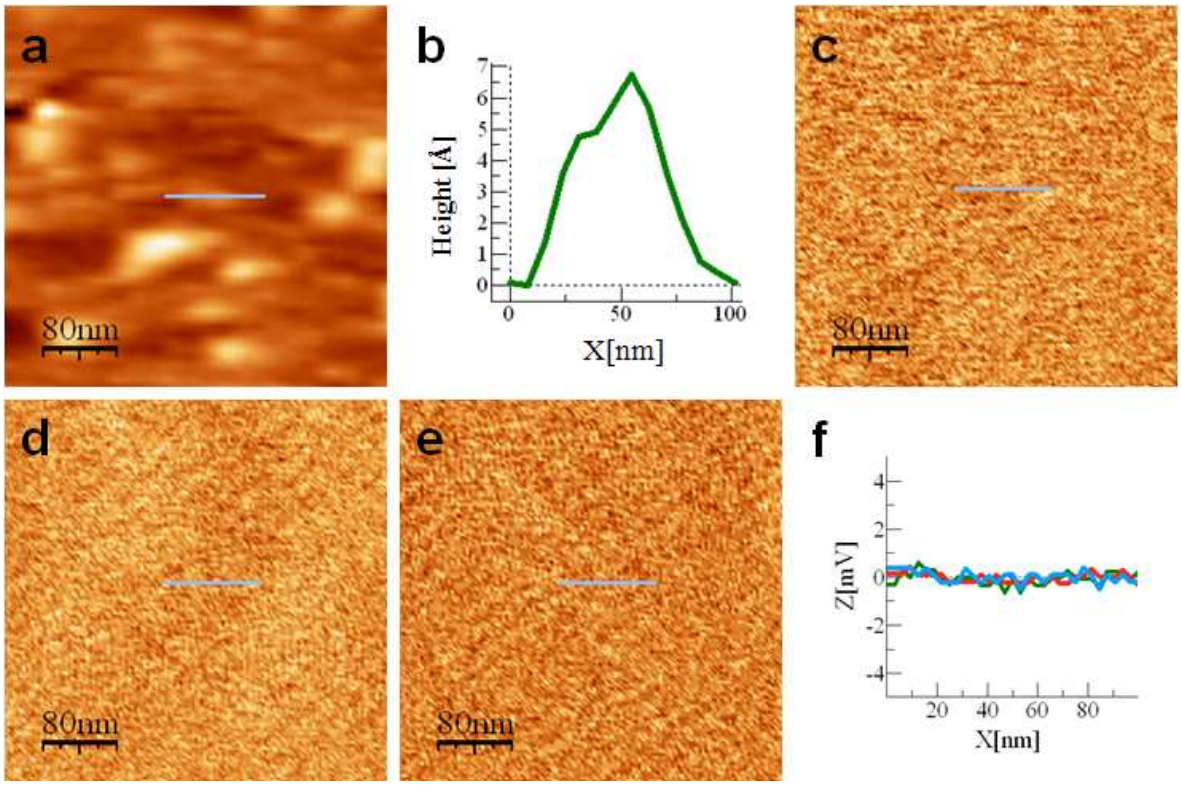

Figure S3: AFM and EFM mesurments of ssDNA. (a) AFM topography image of ssDNA molecules with the same sequence as in the DBCs. (b) Cross section showing the height profile of the ssDNA molecule adsorbed on a mica surface. (c, d, e) Phase shift images (EFM) of the same area at 5 V, 0 V and -5 V, respectively. (f) Line profiles show the magnitude (no observable signal) of the phase shift at the location of the ssDNA (red: 5 V, green: 0 V and blue: -5 V).

AFM and EFM of MWCNTs deposited on a mica surface. Sample preparation: A droplet of 10µL of MWCNT solution (0.1 mg/ml) was applied to a freshly cleaved mica surface for 10 minutes. Then the sample was rinsed gently with triple-distilled water and dried with nitrogen gas. AFM tapping mode images show that the height of the MWCNTs varies along the CNT from 1 to7 nm. As expected, EFM signal was observed at positive and negative bias voltages and no observable signal at zero bias voltage.

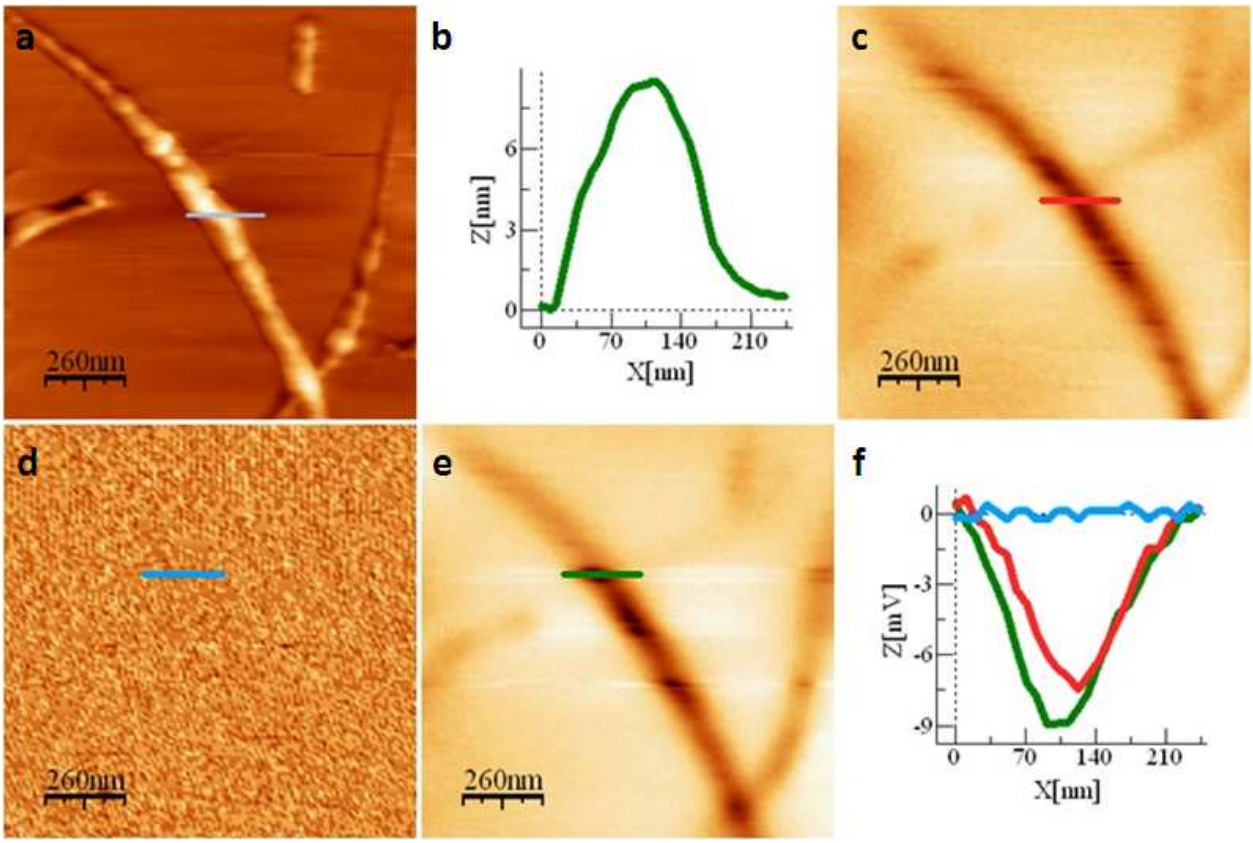

Figure S4 **:** AFM and EFM mesurments of MWCNTs. (a) AFM topography image of MWCNTs. (b) Line profile showing the height of the tube along a cross-section. The value of 6 nm is comparable to the height of the micelles. Phase shift images are shown in (c) +5 V, (d) 0 V and (e) -5 V. (f) A line profile showing the magnitude of the phase shift signal at zero (blue), -5 V (red) and +5 V (green).

Hybridization of DBC micelles with template and bridging ss DNAs

To form lined-up micelle strings, the micelles were hybridized with template (**T1-7**) and bridging (**Bx,y**) ss DNA strands to form linear arrays of particles. The 22 mer ODNs of the corona of the spherical micelles are hybridized to the centers of seven different 42 mer template DNA strands that possess the same complementary sequence in the middle. Each template consists of three parts: one arbitrary 10 mer sequence at the 5'-end (e.g. R_1 in **T1**), followed by the complementary 22 mer sequence in the middle (X', bold black), and finally another arbitary 10 mer. (e.g. R_2 in **T1**). For each template, a bridging 20 mer ss DNA is

designed to interconnect it with a neighboring template sequence by encoding ss DNA complementary to the corresponding arbitrary 10mer; **B1,2** is interconnecting **T1** and **T2** templates. This hybridization resulted in linear particle strings The sequences of the templates and bridging DNAs are shown in Table S1.

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Table S1: The sequences of DNA-*b*-PS, templates, and brindging DNAs. Each color represents complementarity between the template and the bridge.

DNA-*b*-PS: X polymer-5′-**CCTCGCTCTGCTAATCCTGTTA**-3′
The template DNAs (green in Figure 1d): **T1**: R_1-X'-R_2 5′-CATATGAGTA**TAACAGGATTAGCAGAGCGAGG**TTAGAGGCAA-3′ **T2**: R_3-X'-R_4 5′-GTTACAGGAA**TAACAGGATTAGCAGAGCGAGG**GGACCCGAGA-3′ **T3**: R_5-X'-R_6 5′-CTACAAGGGC**TAACAGGATTAGCAGAGCGAGG**AACCGGACCG-3′ **T4**: R_7-X'-R_8 5′-CTAGCCCATA**TAACAGGATTAGCAGAGCGAGG**ACAAGAGAGT-3′ **T5**: R_9-X'-R_{10} 5′-AAGTGGAGCC**TAACAGGATTAGCAGAGCGAGG**AGCTGAGGAA-3′ **T6**: R_{11}-X'-R_{12} 5′-ACAGAGGATC**TAACAGGATTAGCAGAGCGAGG**TAAGCTAAGC-3′ **T7**: R_{13}-X'-R_{14} 5′-CCAGGCGGAG**TAACAGGATTAGCAGAGCGAGG**CGTATAACCC-3′
The bridging DNAs (blue in Figure 1d): **B1,2**: R_3'-R_2' 5′-TTCCTGTAACTTGCCTCTAA-3′ **B2,3**: R_5'-R_4' 5′-GCCCTTGTAGTCTCGGGTCC-3′ **B3,4**: R_7'-R_6' 5′-TATGGGCTAGCGGTCCGGTT-3′ **B4,5**: R_9'-R_8' 5′-GGCTCCACTTACTCTCTTGT-3′ **B5,6**: R_{11}'-R_{10}' 5′-GATCCTCTGTTTCCTCAGCT-3′ **B6,7**: R_{13}'-R_{12}' 5′-CTCCGCCTGGGCTTAGCTTA-3′ **B7,1**: R_1'-R_{12}' 5′-TACTCATATGGGGTTATACG-3′

A solution of all ODNs (seven templates and seven bridges) was prepared by adding 1 µl of each ODN solution with a concentration of 100 µM to 1 mL of ultra pure water. A final mixture of the ODNs and the DBC micelles was prepared by adding 10 µl of the ODN solution and 5 µl of the DBC micelle solution (concentration of 100 µM) to a 100 µl of ultra pure water. This mixture was incubated in a water bath (95°C) for 5 minutes, then allowed to cool down slowly (2 h) to room temperature.